IRRADIATION CREEP AND MECHANICAL PROPERTIES OF TWO FERRITIC-MARTENSITIC STEELS IRRADIATED IN THE BN-350 FAST REACTOR – S.I. Porollo, Yu. V. Konobeev, and A. M. Dvoriashin (State Scientific Center of Russian Federation, (The Institute of Physics and Power Engineering, 249020 Obninsk, Russia) N. I. Budylkin, E. G. Mironova, M. V. Leontyeva-Smirnova, and A. G. Loltukhovsky (State Scientific Center of Russian Federation, A. A. Bochvar All-Russia Research Institute of Inorganic Materials (VNIINM), Moscow, Russia) and F. A. Garner (Pacific Northwest National Laboratory)\*

### **OBJECTIVE**

The objective of this effort is to determine the behavior of several ferritic-martensitic steels when exposed to high fluence neutron irradiation, and to compare the results with those of similar Western steels.

### SUMMARY

Russian ferritic/martensitic steels EP-450 and EP-823 were irradiated to 20-60 dpa in the BN-350 fast reactor in the form of pressurized creep tubes and small rings used for mechanical property tests. Data derived from these steels serves to enhance our understanding of the general behavior of this class of steels. It appears that these steels exhibit behavior that is very consistent with that of Western steels. Swelling is relatively low at high neutron exposure and confined to temperatures <420°C, but may be camouflaged somewhat by precipitation-related densification. The irradiation creep studies confirm that the creep compliance of F/M steels is about one-half that of austenitic steels, and that the loss of strength at test temperatures above 500°C is a problem generic to all F/M steels. This conclusion is supported by post-irradiation measurement of short-term mechanical properties. At temperatures below 500°C both steels retain their high strength ( $\sigma_{0.2}$ =550-600 MPa), but at higher test temperatures a sharp decrease of strength properties occurs. However, the irradiated steels still retain high post-irradiation ductility at test temperatures in the range of 20-700°C.

# **Introduction**

Ferritic-martensitic (F/M) steels are widely used as structural materials of various fission reactors in Russia and other former Soviet states. In particular, EP-450 steel is the reference structural material for hexagonal wrappers of the BN-600 fast reactor subassemblies. The main advantages of F/M steels are their high resistance to swelling, low rate of irradiation creep and rather low activation [1, 2]. The high irradiation resistance of F/M steels therefore encourages their use as structural materials for fusion reactors.

In this paper the results are presented of irradiation creep studies and short-term mechanical properties of two F/M steels, EP-450 (0.12C-13Cr-2MoVNbB) and EP-823 (0.16C-12Cr-MoWSiVNbB), irradiated up to 60 dpa in the BN-350 fast reactor at temperatures in the range 390-520°C. Data derived from these steels also serves to enhance our understanding of the general behavior of this class of steels.

## **Experimental Details**

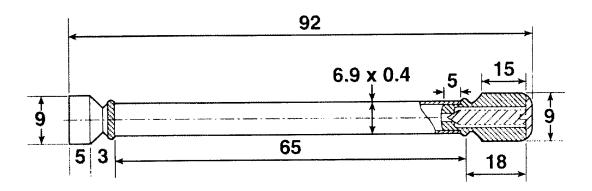
The chemical composition and final heat treatment of thin-wall cladding tubes (6.9 mm outer diameter, 0.4 mm wall thickness) made from EP-450 and EP-823 F/M steels are shown in Table 1. Two types of samples were constructed from these tubes and were used in the experiment: pressurized creep tubes (see Figure 1), and ring specimens of 2 mm in length cut from the tubes for measurement of mechanical properties. The creep tubes were filled with argon of 99.998% purity through a needle valve located in the large plug end to produce hoop stresses  $\sigma_h$  ranging from 0 to 294 MPa at the irradiation temperature.

<sup>\*</sup> Pacific Northwest National Laboratory (PNNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

Steel	Content, wt %														
2.001	С	Si	Mn	S	Р	Cr	Ni	Мо	Nb	Ti	Al	V	В	N <sub>2</sub>	Others
EP-	0.14	0.20	0.31	0.009	0.017	12.95	0.20	1.54	0.47	-	-	0.22	0.004	-	-
450	Heat treatment: solution treated 1050°C, 1s + aged 850°C, 5s.														
EP-	0.18	1.05	0.60	0.008	0.012	11.40	0.70	0.67	0.20	0.03	0.0	3 0.40	0.004	0.0	4 W=
823															0.65
															Ce=
															0.01
İ		Heat treatment: normalization 1050°C, 15 min + tempering 740°C, 1 h													

Table 1. Chemical composition of EP-450 and EP-823 F/M steels.

Figure 1. Argon-pressurized creep tubes irradiated in BN-350 reactor. All dimensions are given in mm.



The creep tubes were irradiated in the BN-350 fast reactor (Kazakhstan) for 9430 h in two experimental assemblies located in the fifth row of the low enrichment zone. Each of the two assemblies operated over a different temperature range (390-410 and 480-520°C), with the temperature calculated based on prior measurements. At the core midplane this corresponds to a maximum total neutron fluence of  $1.9\times10^{23}$  n/cm² ( $1.36\times10^{23}$  n/cm² (E>0.1MeV)) or 60 dpa (NRT) for both assemblies. The tubes were placed in perforated cylindrical baskets of 76 mm diameter in direct contact with flowing sodium. In both assemblies the EP-450 tubes were located at three axial levels: basket #4 at 0 to +100 mm, basket #10 at +300 to +400 mm, and basket #15 of +650 to +750 mm, all measured from the core midplane. EP-823 tubes were located only in basket #4 of both assemblies, reaching 380 and 490°C.

Using short fuel pins placed in the bottom part of the assemblies allowed heating of the baskets to their operating temperature. The irradiation conditions for each basket are shown in Table 2. Concurrent with the reactor tests, several tubes fabricated from EP-450 and EP-823 steels were tested outside the reactor for 9800 h at temperatures of 400°C and 500°C and at the same levels of hoop stress.

basket #	4	10	15		
assembly #1					
T <sub>irr.</sub> , °C	390±10	400±10	410±10		
assembly #2					
T <sub>irr.</sub> , °C	480±10	500±10	520±10		
φt, 10 <sup>23</sup> n/cm <sup>2</sup> E>0	1.90	1.42	0.60		
φt, 10 <sup>23</sup> n/cm <sup>2</sup> E>0.1 MeV	1.36	1.02	0.45		
dpa	60	45	20		
σ <sub>h</sub> , MPa	0; 98; 196; 294	0; 98; 196; 294	0; 98; 196; 294		

Table 2. Irradiation conditions for creep tubes in the BN-350 fast reactor.

Ring specimens of 2 mm length for measurement of mechanical properties were irradiated in flowing sodium at 490±10°C to 50 dpa in only one assembly in basket #9, located at +260+300 mm. After irradiation, the tube surfaces were cleaned in 50% ethanol—water solution and the diameter was measured using a micrometer. The diameter of each creep tube was measured at the tube middle and also at the distance of 15 mm from each end of the tube for two orientations differing by a 90° rotation around the tube axis. The irradiation creep strain  $\epsilon^{ic}$  was determined as the difference between the total diametral strain and the diametral strain due to swelling, determined by the strain of stress-free tubes. The irradiation creep modulus  $B_{ic}$  was determined from the following expression:

$$B_{ic} = \varepsilon^{ic}/0.75\sigma_h(dpa)$$
.

It should be noted that this modulus is a composite creep rate, containing transient creep, creep in the absence of swelling (defined as the creep compliance  $B_0$ ) and the swelling–enhanced creep component, usually defined as  $DS_{dot}$ , where D is the creep-swelling coupling coefficient, and  $S_{dot}$  is the swelling rate. Unfortunately, there may also be some precipitation-related strains included as well.

The mechanical characteristics for both irradiated and non-irradiated ring specimens were measured using a shielded tensile testing machine at the test temperatures ranging from 20 to 700°C. The aging time at given temperature was 10 min for unirradiated specimens and 20 min for irradiated ones. Two or three separate specimens were tested at a given temperature.

#### Results

The thermal creep tests of surveillance tubes at 500°C for 9800 hours demonstrated that thermal creep strains in EP-450 could reach several percent, while those at 400°C were much smaller.

All irradiated creep tubes had no visible defects and were still gas-tight. The measured total diametral strain, irradiation creep strain and the irradiation creep moduli for EP-450 and EP-823 steels are shown in Tables 3 and 4. At 390°C the creep characteristics for these steels are essentially equal. At 480°C the irradiation creep strain is distinctly larger in EP-823. In Table 3 it can be seen that stress-free swelling of EP-450 has two possible temperature maxima, located at ~400°C and ~500°C. Interestingly, two temperature maxima, located at ~425°C and ~510°C have been observed in Ref. [3] for pure iron irradiated in DFR to 30 and 23 dpa at a dose rate higher by one order of magnitude.

Table 3. Irradiation creep characteristics for EP-450 steel.

$\sigma_h$ ,	390°C/60 dpa		dpa	4	400°C/4	l5 dpa	410°C/20 dpa			
MPa	Δd/d,	ε <sub>ic.</sub> ,	B <sub>ic</sub> , 10 <sup>-6</sup>	$\Delta d/d$ $\epsilon_{ic.}$ , $B_{ic}$ , $10^{-2}$		B <sub>ic</sub> , 10 <sup>-6</sup>	∆d/d	$\epsilon_{\sf ic},$	B <sub>ic</sub> , 10 <sup>-6</sup>	
	%	%	Mpa <sup>-1</sup>	%	%	MPa <sup>-1</sup>	%	%	MPa <sup>-1</sup>	
			×dpa <sup>-1</sup>			×dpa <sup>-1</sup>			×dpa <sup>-1</sup>	
0	0.26	0	0	0.41	0	0	0.14	0	0	
98	0.59	0.33	0.7	0.43	0.02	0.06	0.14	0	0	
196	1.25	0.99	1.1	0.76	0.35	0.5	0.34	0.20	0.3	
294	1.77	1.51	1.1	1.05	0.64	0.6	0.40	0.26	0.3	
$\sigma_{h},$	480°C/60 dpa			500°C/45 dpa			520°C/20 dpa			
MPa	Δd/d,	ε <sub>ic</sub> ,	B <sub>ic</sub> , 10 <sup>-6</sup>	∆d/d	$\epsilon_{\sf ic},$	B <sub>ic</sub> , 10 <sup>-6</sup>	∆d/d	$\epsilon_{\sf ic},$	B <sub>ic</sub> , 10 <sup>-6</sup>	
	%	%	MPa <sup>-1</sup>	%	%	MPa <sup>-1</sup>	%	%	MPa <sup>-1</sup>	
			×dpa <sup>-1</sup>			×dpa <sup>-1</sup>			×dpa <sup>-1</sup>	
0	0	0	0	0.22	0	0	0	0	0	
98	0	0	0	0.29	0.07	0.2	0.29	0.29	2.0	
196	0.29	0.29	0.3	0.36	0.14	0.2	0.36	0.36	1.2	
294	0.58	0.58	0.4	0.94	0.72	0.7	0.58	0.58	1.3	

Table 4. Irradiation creep characteristics for EP-823 steel.

	390°C/60 dpa						
$\sigma_h$ ,	∆d/d,%	ε <sub>i.c.</sub> ,	B <sub>ic</sub> , 10 <sup>-6</sup>				
MPa		%	MPa⁻¹				
			×dpa <sup>-1</sup>				
0	0.20	-	-				
98	0.64	0.44	1.0				
196	0.92	0.72	0.8				
294	1.08	0.88	0.7				
	480°C/60 dpa						
$\sigma_h$ ,	$\Delta d/d,\%$	ε <sub>i.c.</sub> ,%	B <sub>ic</sub> , 10 <sup>-6</sup>				
MPa			Mpa <sup>-1</sup>				
			×dpa <sup>-1</sup>				
0	0	0	0				
98	0.09	0.09	0.2				
196	0.14	0.14	0.2				
294	1.85	1.85	1.4				

The mechanical characteristics of EP-450 and EP-823 steels, both before and after neutron irradiation at 490±10°C to 50 dpa, are shown in Figures 2-5. The strength properties of EP-450 before and after irradiation at 490°C to 50 dpa do not differ significantly, and radiation-induced loss of elongation is only significant at higher test temperatures (Figures 2, 3). Neutron irradiation of EP-823 steel resulted in significantly more hardening and somewhat greater loss of ductility across a wider temperature range (Figures 4, 5). At test temperatures above 500°C a sharp decrease of strength was observed for these two steels in both the initial and irradiated conditions.

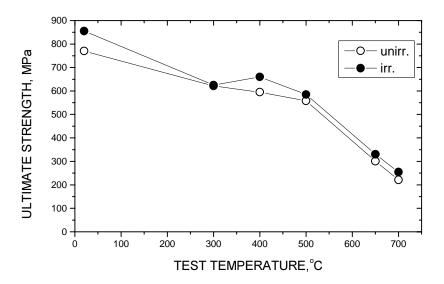


Figure 2. Ultimate strength for the EP-450 type steel as a function of test temperature after irradiation to 50 dpa at  $490^{\circ}$ C.

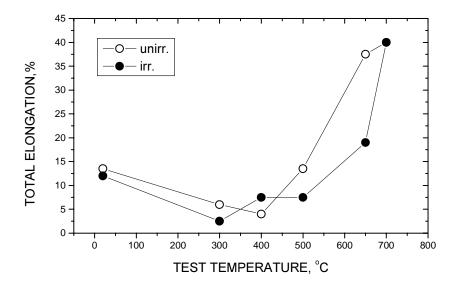


Figure 3. Total elongation for the EP-450 type steel as a function of test temperature after irradiation to 50 dpa at 490°C.

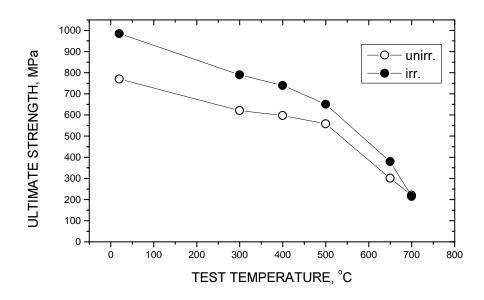


Figure 4. Ultimate strength for the EP-823 type steel as a function of test temperature after irradiation to 50 dpa at  $490^{\circ}\text{C}$ 

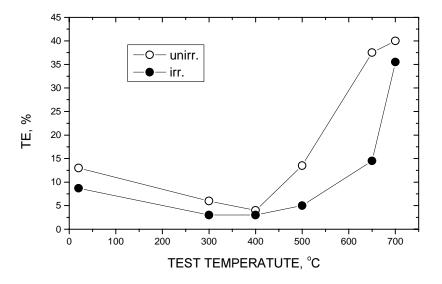


Figure 5. Total elongation for the EP-823 type steel as a function of test temperature after irradiation to 50 dpa at 490°C.

# **Discussion**

Swelling of EP-450, as indicated by positive strains in the diameter of the unstressed tubes, appears only in the 390-420°C range, consistent with the reported behavior of other F/M steels. The maximum swelling appears to be ~1.2% at 400°C and 45 dpa, providing that the zero-stress strains do not include any contributions from precipitation. In the more limited

irradiation matrix of EP-823, there also appears to be some swelling on the order of  $\sim$ 0.6% at 400°C, but not at 500°C.

The temperature and stress dependencies of the irradiation creep modulus of EP-450 steel are shown in Figure 6. The rather low modulus observed at 98 MPa appears to signal the presence of negative strains arising from precipitate-related densification, also observed in some other F/M steels. It is also seen that at hoop stresses of 98, 196 and 294 MPa the modulus exhibits a flat minimum within the temperature range 410-480°C, with the modulus not exceeding  $0.4\times10^{-6}$  (MPa×dpa)<sup>-1</sup> at the higher stress levels in this temperature range. There appears to be no significant swelling at these temperatures.

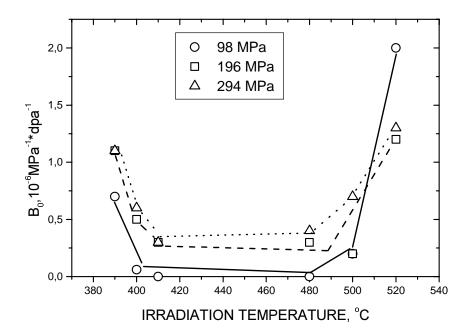


Figure 6. Creep compliance for the EP-450 type steel versus the irradiation temperature for three stress levels.

This low modulus value is one-third to one-half that routinely observed in austenitic steels (4) and is also consistent with moduli measured on other F/M steels in the absence of swelling, as summarized by Toloczko and Garner, who note that the creep compliance  $B_0$  is on the order of  $0.5\times10^{-6}$  (MPa×dpa)<sup>-1</sup> in HT9, 9Cr-1Mo and other F/M steels, especially when densification is not operating (5-9).

The observation of moduli larger than this value at temperatures below 410°C reflects the onset of the  $DS_{dot}$  contribution of irradiation creep. Since swelling in F/M steels has been observed to be enhanced by applied stress (5), it is assumed that a similar stress-dependency may be operating in this experiment. When added to the apparent densification proceeding in this steel, this complicates a separation of the  $B_0$  and  $DS_{dot}$  contributions. Toloczko and Garner have earlier shown that the creep-swelling coupling coefficient of F/M and austenitic steels are essentially equal, unlike the difference observed in  $B_0$  (7).

It is thought to be particularly significant that the EP-450 and EP-823 steels exhibited very consistent creep behavior at both 390 and 500°C, even though there are substantial differences in composition. This tends to confirm once again that the B<sub>0</sub> component of

irradiation creep is relatively insensitive to composition and starting state within a given alloy class, as observed in both austenitic and F/M steels (4, 8, 9).

With increasing irradiation temperature to 520°C the irradiation creep modulus EP-450 increases to 2×10<sup>-6</sup> (MPa×dpa)<sup>-1</sup>. A similar temperature dependence of the irradiation creep modulus of EP-450 steel has been found in measurements of hexagonal wrappers of sub-assemblies in BN-600 fast reactor [10] and in the studies of Toloczko et al. on HT9 (8,9). An increase of the irradiation creep modulus of EP-450 at irradiation temperatures higher than 500°C is thought to be due to decreases in its strength characteristics in this temperature region, and the onset of greater-than-linear creep, as was also observed in HT9 (8,9). The relatively large strains observed at 500°C in the thermal control specimens supports this interpretation.

Measurements of short-term mechanical properties of EP-450 steel have revealed that irradiation hardening of this steel is relatively insignificant at the irradiation temperature of 500°C. This observation is in agreement with data obtained earlier in examination of EP-450 fuel pin cladding after irradiation in BN-350 [11]. For EP-823 steel the radiation-induced increase of ultimate strength is slightly higher (225 MPa). With increasing test temperature a sharp decrease of ultimate strength from 600 MPa at 500°C to 200 MPa at 700°C is observed in both steels. Similar behavior is observed in most other F/M steels.

#### Conclusions

The Russian ferritic/martensitic steels EP-450 and EP-823 have served as structural components in various fission reactors and may serve in future fusion devices. Data derived from these steels also serves to enhance our understanding of the general behavior of this class of steels. It appears that these steels exhibit behavior that is very consistent with that from Western steels. Swelling is relatively low at high neutron exposure and confined to temperatures <420°C, but may be camouflaged somewhat by precipitation-related densification. These irradiation creep studies confirm that the creep compliance of F/M steels is about one-half that of austenitic steels, and that the loss of strength at test temperatures above 500°C is a problem generic to all F/M steels.

Measurements of short-term mechanical properties for irradiated steels have shown, that at test temperatures below 500°C both steels retain their high strength ( $\sigma_{0,2}$ =550-600 MPa), but at higher test temperatures a sharp decrease of strength properties occurs. However, the irradiated steels still retain high ductility at test temperatures in the range of 20-700°C.

## **ACKNOWLEDGEMENTS**

The Civilian Research and Development Foundation under Grant RE2-2224 sponsored the Russian portion of this work. The Office of Fusion, U.S. Department of Energy provided support for the U.S. portion of this research under contract DE-AC06-76RLO 1830 with Battelle Memorial Institute.

# REFERENCES

- [1] V. S. Khabarov, V. D. Dmitriev, A. M. Dvoriashin, V. V. Romaneev, E. A. Medvedeva, "Mechanical properties and microstructure of neutron-irradiated ferritic-martensitic steel, used as wrapper material for the BN-350 and BN-600 fast reactor", Proceedings of International Conference" Fast reactor core and fuel structural behavior", Inverness, 4-6 June 1990, p.p. 263-267.
- [2] A. G. Ioltukhovski, et. al., "Material science and manufacturing of heat-resistant reduced-activation ferritic-martensitic steels for fusion", Journal of Nuclear Materials, 283-287 (2000), 652-656.

- [3] E. A. Little and D. A. Stow "Void swelling in iron and ferritic steels. II. An experimental survey", Journal of Nuclear Materials, 1973, Vol. 87, No. 1, pp. 25-39.
- [4] F. A. Garner, Chapter 6: "Irradiation Performance of Cladding and Structural Steels in Liquid Metal Reactors," Vol. 10A of Materials Science and Technology: A Comprehensive Treatment, VCH Publishers, 1994, pp. 419-543.
- [5] M. B. Toloczko, F. A. Garner, and C. R. Eiholzer, "Irradiation Creep and Swelling of the U.S. Fusion Heats of HT9 and 9Cr-1Mo to 208 dpa at ~400°C," J. of Nucl. Mater., 212-215 (1994) 604-607.
- [6] M. B. Toloczko and F. A. Garner, "Irradiation Creep and Void Swelling of Two LMR Heats of HT9 at ~400°C and 165 dpa", J. of Nucl. Mater., 233-237 (1996) 289-292.
- [7] M. B. Toloczko and F. A. Garner, "Variability of Irradiation Creep and Swelling of HT9 Irradiated to High Neutron Fluence at 400-600°C", Effects of Radiation on Materials: 18th International Symposium, ASTM STP 1325, R. K. Nanstad, M.. Hamilton, F. A. Garner and A. S. Kumar, Eds., American Society of testing and Materials, 1999, pp.765-779.
- [8] M. B. Toloczko, F. A. Garner and C. R. Eiholzer, "Irradiation Creep of Various Ferritic Alloys Irradiated at ~400°C in the PFR and FFTF Reactors", J. of Nucl. Mater., 258-263 (1998), pp. 1163-1166.
- [9] M. B. Toloczko, B. R. Grambau, F. A. Garner and K. Abe, "Comparison of Thermal Creep and Irradiation Creep of HT9 Pressurized Tubes at Test Temperatures from ~490°C to 600°C", Proceedings of Symposium on Effects of Radiation on Materials, 20th International Symposium, ASTM STP 1405, S. T. Rosinski, M. L. Grossbeck, T. R. Allen, and A. S. Kumar, Eds., American Society for Testing and Materials, West Conshohocken, PA, 2002, pp. pp. 557-569.
- [10] L. V. Zabudko, I. N. Kravchenko, O. S. Korostin, A. N. Ogorodov, "Analysis of the BN-600 reactor subassemblies operating experience", Proc. of Inter. Conf. "Fast reactor core and fuel structural behavior", Inverness, 4-6 June 1990, p.p. 289-292.
- [11] A. M. Dvoriashin, V. D. Dmitriev, V. S. Khabarov, "The Effect of Neutron Irradiation on the Microstructure and Tensile Properties of 1Cr13Mo2NbVB Steel", Effects of Radiation on Materials: 15th International Symposium, ASTM STP 1125, R. E. Stoller, A. S. Kumar, and D. S. Gelles, Eds., American Society for Testing and Materials, Philadelphia, 1992, pp. 1180-1189.